

Polarization dynamics in quantum dots: The role of dark excitons.

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Abstract

We study an impact of the fine structure of the heavy-hole ground state exciton confined in semiconductor quantum dots on the photoluminescence polarization dynamics solving the relevant system of the rate equations. The presence of the dark excitons is usually ignored and the polarization decay is assumed to be caused by direct transitions within the radiative doublet. We demonstrate that in strongly confined quantum dots the dark excitons, which are energetically well below the bright excitons, have actually a decisive effect on the polarization dynamics due to their persistent nature. The linear polarization shows nonexponential decay controlled by a conversion of the dark into a bright exciton. To get quantitative answers for specific quantum dot structures, all the necessary information can be obtained already from experiments on the luminescence dynamics following nonresonant excitation in these dots.

Keywords:

quantum dots; excitons; spin relaxation; photoluminescence

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I. INTRODUCTION

The dynamics of the spin-polarized excitons in semiconductor quantum dots (QDs) attracted a great attention from both fundamental and technological point of view during the two previous decades. The spin relaxation of excitons between the bright and dark states strongly limits the generation of single photons from the dots[1–3], whereas the spin-flip transitions within the bright exciton doublet, so-called longitudinal spin relaxation, limit the performance of entangled photon pair generators[4, 5]. So according to the numerous time-resolved experiments, an inclusion of dark excitons to the relaxation processes essentially modifies the luminescence dynamics following nonresonant excitation, whereas the longitudinal spin relaxation leads to a decay of the luminescence polarization. Theoretical examinations identified two relevant microscopic mechanisms of an intrinsic nature responsible for thermally activated spin-flip transitions within the ground state of the heavy-hole exciton confined in a QD. The first mechanism is mediated by the exchange interaction[6–8] and the second mechanism is governed by the spin-orbit coupling[9]. For self-assembled QDs in vertical field effect structures, a Kondo-like interaction between a localized electron in a dot and a delocalized electron in the back contact was proposed as the main microscopic mechanism of the dark-to-bright spin flip in Ref.[10]. Recently, an additional depolarization source due to transitions between the bright states via the dark states has been identified in Ref.[11], so that the dark-to-bright transitions also can limit the performance of entangled photon pair generators. An impact of this relaxation channel on the polarization dynamics, however, was still not examined in details. The only exception here, as far as we know, is the theoretical paper[8], where the reported results are based on the numerical calculations for the specific case of the InAs QDs.

In this paper we present the analytical results on the polarization dynamics in a single QD solving the phenomenological rate equations similar to those considered in Ref.[8]. The paper is organized as follows. Section II contains the problem statement and the obtained analytical results are discussed in section III. In section IV the quantitative estimation of depolarization effects in various quantum dots studied recently in nonresonantly excited photoluminescence experiments [10, 12–14] and a summary as well are given.

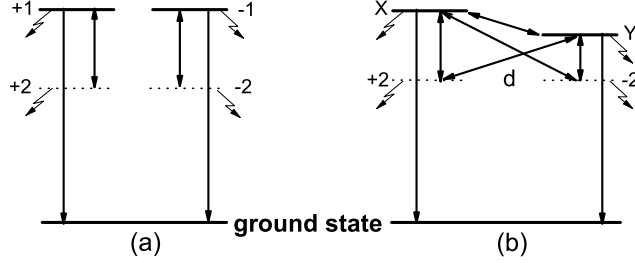


FIG. 1: Diagram of transitions included in (a) a three-level model and (b) a four-level model.

II. FOUR-LEVEL MODEL.

Many experimental works devoted to the exciton dynamics in single quantum dots report on a biexponential decay of the band edge exciton luminescence as observed e.g. in CdSe/ZnS QDs in Ref.[15]. The results are interpreted satisfactory in terms of a three-level model of a zero exciton ground level and two mixed fine structure states of the heavy-hole exciton, the bright state $|+1\rangle$ ($|-1\rangle$) and the dark state $|+2\rangle$ ($|-2\rangle$), see Fig.1(a). This simple model, however, contains no interplay between two bright states (spin doublet), which are assumed to be coupled to the dark states in pairs, and therefore is not able to describe the impact of the longitudinal spin relaxation. In fact, because of the reduced symmetry and anisotropic exchange interaction, the two bright states are separated in energy and form two linearly and orthogonally polarized eigenstates $|X\rangle$ and $|Y\rangle$. The spin-flip transitions $|X\rangle \leftrightarrow |Y\rangle$ between these states directly evidence the longitudinal exciton spin relaxation. A coupling of *both* bright states to *both* dark states opens an additional channel due to the sequential process like as $|X\rangle \rightarrow |d\rangle \rightarrow |Y\rangle$ [8, 11].

Based on this scenario we consider a four level model system containing two bright states $|X\rangle$ and $|Y\rangle$, a two-fold degenerate dark state [16], and the crystal ground state, as is shown in Fig.1(b). We assume that a short laser pulse leads to a partial occupation of the involved excitonic states. Let $N_X(0)$ and $N_Y(0)$ be the population probabilities (occupations) of the bright states at the time of initialization $t = 0$ and $N_d(0)$ is the joint initial occupation of the dark state. Supposing that both bright states are coupled to both dark states [11],

further temporal evolution of the occupations N_X, N_Y and N_d is described by the following rate equations

$$\left. \begin{aligned} \dot{N}_X(t) &= -(\tilde{\gamma}_r + 2\gamma_{Xd} + \gamma_{XY})N_X(t) + \gamma_{YX}N_Y(t) + \gamma_{dX}N_d(t), \\ \dot{N}_Y(t) &= -(\tilde{\gamma}_r + 2\gamma_{Yd} + \gamma_{YX})N_Y(t) + \gamma_{XY}N_X(t) + \gamma_{dY}N_d(t), \\ \dot{N}_d(t) &= -(\gamma_{nr} + \gamma_{dX} + \gamma_{dY})N_d(t) + 2\gamma_{Xd}N_X(t) + 2\gamma_{Yd}N_Y(t), \end{aligned} \right\} \quad (1)$$

where $\tilde{\gamma}_r = \gamma_r + \gamma_{nr}$, γ_r is the radiative recombination rate for the bright exciton and γ_{nr} simulates any nonradiative loss (usually the same for both the bright exciton and the dark exciton). The rates γ_{id} and γ_{di} , $i = X, Y$, characterize the transitions between the bright and dark exciton states and γ_{XY}, γ_{YX} are the transition rates between the bright states. For the dark exciton, which is optically forbidden (in the dipole approximation), the radiative recombination rate is taken to be zero. As noted above, a similar system of the rate equations, but with $\gamma_{nr} = 0$, was calculated numerically for a specific case in Ref.[8].

Below we will restrict our consideration by strongly confined QDs (like e.g. self-assemble InGaAs/GaAs QDs) for which the reported values of the bright-dark splitting Δ_0 [17] are typically (at least) a few times larger than those for the anisotropic splitting Δ_{XY} between the bright states [14]. One can assume therefore that the spin-flip rates for both bright excitons must be close in magnitude and further simplification of the above system is possible. In what follows we take $\gamma_{Xd} = \gamma_{Yd} \equiv \gamma_{bd}/2$ and, similarly, $\gamma_{dX} = \gamma_{dY} \equiv \gamma_{db}/2$. Excluding also a very low temperature range, where $kT \ll \Delta_{XY}$, we can take $\gamma_{XY} = \gamma_{YX} \equiv \gamma_L/2$. Finally, it is convenient to replace the occupations N_X and N_Y by the joint occupation $N_+ = N_X + N_Y$ and the occupation difference $N_- = N_X - N_Y$. Under the above approximations the equation system (1) splits into the equation for $N_-(t)$ and two coupled equations for $N_+(t)$ and $N_d(t)$:

$$\left. \begin{aligned} \dot{N}_-(t) &= -(\tilde{\gamma}_r + \gamma_{bd} + \gamma_L)N_-(t), \\ \dot{N}_+(t) &= -(\tilde{\gamma}_r + \gamma_{bd})N_+(t) + \gamma_{db}N_d(t), \\ \dot{N}_d(t) &= -(\gamma_{nr} + \gamma_{db})N_d(t) + \gamma_{bd}N_+(t). \end{aligned} \right\} \quad (2)$$

Solving the above equations, for the occupations of the bright states we get

$$N_{X,Y}(t) = F_1 e^{-\gamma_1 t} + F_2 e^{-\gamma_2 t} \pm F_3 e^{-(\tilde{\gamma}_r + \gamma_{bd} + \gamma_L)t}. \quad (3)$$

Here the decay rate $\gamma_{1,2} = 0.5[(\gamma_r + 2\gamma_{nr} + \gamma_{bd} + \gamma_{db}) \pm \delta]$ with $\delta = \sqrt{\Gamma^2 + 4\gamma_{db}\gamma_{bd}}$ and $\Gamma = \gamma_r + \gamma_{bd} - \gamma_{db}$. The amplitudes $F_{1,2} = [(\delta \pm \Gamma) N_+(0) \mp 2\gamma_{db} N_d(0)]/4\delta$ and $F_3 =$

$N_-(0)/2$. Similarly, the occupations of the dark states $N_d(t) = D_1 e^{-\gamma_1 t} + D_2 e^{-\gamma_2 t}$ with $D_{1,2} = [(\delta \mp \Gamma) N_d(0) \mp 2\gamma_{bd} N_+(0)]/4\delta$.

The above results have a simple interpretation. **(i)** Let all involved excitonic states are initially equally populated - the situation realized at nonresonant excitation. Then the difference $N_-(0) = 0$ and the initial condition $N_X(0) = N_Y(0)$ persists over time, that is both radiative excitons decay identically and show the two-component structure - the result which is typical for a three level model. **(ii)** Let now only one bright state be initially populated, that is $N_-(0) = 1$ or $N_-(0) = -1$. This situation is realized at resonant (linearly polarized) excitation. In this case the amplitude of the third component for N_X and N_Y differs from zero, so that the initially established population is redistributed over time within the fine-structure levels. This ultimately induces a depolarization of the resulting emission. The degree of the linear polarization of the emission is directly related to the occupations by $P(t) = N_-(t)/N_+(t)$.

III. DEGREE OF LINEAR POLARIZATION

Let us now consider a short pulsed laser excitation at $t = 0$ leading to a population of one of the bright states, say the $|X\rangle$ state, so that $N_X(0) = 1$, but $N_Y(0) = N_d(0) = 0$. According to result (3), the degree of linear polarization of the emitted signal is given by

$$P(t) = \frac{2\delta e^{-\frac{1}{2}(\Gamma+2\gamma_L)t}}{(\delta + \Gamma) e^{-\delta t/2} + (\delta - \Gamma) e^{\delta t/2}}. \quad (4)$$

The obtained polarization decay is entirely caused by fine-structure effects. Sole direct spin-flip transitions $|X\rangle \leftrightarrow |Y\rangle$ within the radiative doublet lead to a partial redistribution of the occupation between these states, but the dark exciton states are, evidently, unaffected. This is the familiar case of an exponentially decaying polarization $P(t) = e^{-\gamma_L t}$. The sequential process like as $|X\rangle \rightarrow |d\rangle \rightarrow |Y\rangle$, however, causes a nonexponential decay of the joint occupation of the bright states and effectively contributes to a decay of the linear polarization. This fact is clearly reflected in the time-integrated polarization for which we get

$$\overline{P} = \frac{\tilde{\gamma}_r + \gamma_{bd} - \gamma_L^*}{\tilde{\gamma}_r + \gamma_{bd} + \gamma_L}. \quad (5)$$

Here the involved quantity $\gamma_L^* = \gamma_{bd}\gamma_{db}(\gamma_{nr} + \gamma_{db})^{-1}$ is interpreted as the rate of the above sequential transition between the bright states. Such a transition can be regarded as a

second-order (or indirect) relaxation process, where the dark state serves as the intermediate state [11]. The sequential spin relaxation is, evidently, the more effective the faster is a conversion of the dark into a bright exciton in comparison with the nonradiative relaxation. In a strong conversion regime, where $\gamma_{db} \gg \gamma_{nr}$, the sequential transition rate reaches the values $\gamma_L^* \simeq \gamma_{bd}$, whereas $\gamma_L^* \ll \gamma_{bd}$ in a weak conversion regime, where $\gamma_{bd} \ll \gamma_{nr}$. Note that a relation between the rates γ_{db} (γ_{bd}) and γ_{nr} can be examined in experiments on the luminescence dynamics following nonresonant excitation [12].

Let us now rewrite the expression (5) for the integrated polarization in a more convenient form $\bar{P} = \tilde{\gamma}_r(\tilde{\gamma}_r + \gamma_s)^{-1}$, where $\gamma_s = \tilde{\gamma}_r(\gamma_L + \gamma_L^*)(\tilde{\gamma}_r + \gamma_{bd} - \gamma_L^*)^{-1}$ can be viewed as an *effective* polarization decay rate. In the special case of no bright-dark coupling the rate γ_s is identical to γ_L , the polarization decays exponentially, and the definition $\bar{P} = \tilde{\gamma}_r/(\tilde{\gamma}_r + \gamma_L)$ is the standard one. The more realistic case of an impact of the dark states complicates the situation. The (effective) decay rate now has an intricate form and, as could be expected, is proportional to the total rate of the longitudinal spin relaxation, $\gamma_s \sim (\gamma_L + \gamma_L^*)$.

For the spin relaxation of an intrinsic nature further simplification of the above results is possible. In this case the spin-flip transitions are thermally activated and the bright exciton relaxes predominantly to the lowest dark state [19]. This means that direct transitions $|X\rangle \leftrightarrow |Y\rangle$ are slow compared to the bright-to-dark relaxation and therefore can be abandoned in the analysis, so that we can chose $\gamma_L = 0$ in the first of the equations (2) and, consequently, in expression (4). An effective decay rate in this case is approximated by

$$\gamma_s = \frac{\tilde{\gamma}_r \gamma_L^*}{\tilde{\gamma}_r + \gamma_{bd} - \gamma_L^*}. \quad (6)$$

It is remarkable that all the dot parameters necessary to estimate depolarization effects in undisturbed QDs can be deduced in nonresonantly excited photoluminescence experiments. It follows immediately from (6) that no noticeable effects have to occur in a weak conversion regime, where all the involved rates γ_{bd} , γ_L^* and γ_s as well are much smaller than γ_{nr} . Respectively, the time-integrated polarization preserves its initial value, $\bar{P} = 1$. In a strong conversion regime, where as we saw above $\gamma_L^* \simeq \gamma_{bd}$, an efficiency of the polarization decay and the actual values of $\bar{P} = \tilde{\gamma}_r(\tilde{\gamma}_r + \gamma_{bd})^{-1}$ as well depend on the ratio $\gamma_{bd}/\tilde{\gamma}_r$. For typical self-assembled QDs at low temperatures, intrinsic spin-flip transitions are much slower than the radiative recombination, as estimated theoretically [8, 11] and observed experimentally, see e.g. [12]. Hence noticeable effects here can be expected only at enough high temperatures.

Note that self-assembled QDs in a vertical field effect structures, similar to those studied in Ref.[10], can also serve as an example of the QDs systems for which the bright-to-dark processes are preferential. A strong dependence of the characteristic rate γ_{db} on a voltage in these dots rules out an intrinsic spin flip mechanism, see discussion below in section IV c.

IV. QUANTITATIVE RESULTS

In the previous section we observed that the experimental examination of the luminescence dynamics following nonresonant excitation in QDs presents sufficient information to make a conclusion about depolarization effects in these dots. It is possible to get quantitative answers on the polarization decay rate in such QDs for which the nonresonant experimental data are at hand and therefore can be used as input. It has to be implemented for many QD systems in the strong confinement limit and below we consider several examples.

a. Self-assembled In(Ga)As QDs. A recent series of works [12] reports on the experimental investigation of nonresonantly excited luminescence dynamics in self-assembled In(Ga)As QD structures. Actually the following characteristic rates were deduced from experiments at 14 K: $\gamma_r \simeq 1 \text{ ns}^{-1}$, $\gamma_{nr} \simeq 0.1 \text{ ns}^{-1}$, and $\gamma_{db} \simeq 0.01 \text{ ns}^{-1}$. Although no data on γ_{bd} and Δ_0 as well are reported, we can consider that $\gamma_{bd} \simeq \gamma_{db}$ (Δ_0 in InAs QDs is typically of a few hundreds of μeV [17], whereas $kT = 1.2 \text{ meV}$ at 14 K) [20]. As a result, the (effective) polarization decay rate is calculated from (6) to be $\gamma_s \simeq 10^{-3} \text{ ns}^{-1}$, so that we can conclude that In(Ga)As QDs examined in Ref.[12] shouldn't show any depolarization effects for the reported experimental conditions. This conclusion is in accordance to earlier experimental results from Ref.[21], where strictly no decay of the linear luminescence polarization was observed in self-organized InAs/GaAs QDs at low temperature.

b. Self-assembled CdSe/ZnSSe/MgS QDs. Recently the high-quality CdSe/ZnSSe/MgS self-assembled QDs were proposed in Ref.[13] as an excellent model system to analyze experimentally the dark-bright interplay in a wide range of temperatures. The (nonresonantly excited) luminescence dynamics investigated in [13] is well explained by a three-level model and no modification of the nonradiative losses was found all the way up to room temperature. The following parameters were deduced from the experiment: $\gamma_r = 1.54 \text{ ns}^{-1}$, $\Delta_0 \simeq 1.8 \text{ meV}$, $\gamma_{bd}(0\text{K}) = \gamma_0 = 0.166 \text{ ns}^{-1}$ [20], and $\gamma_{slow}(7\text{K}) \simeq \gamma_{nr} + \gamma_{db}(7\text{K}) = 0.02 \text{ ns}^{-1}$ (the rate of the slow luminescence component at 7 K).

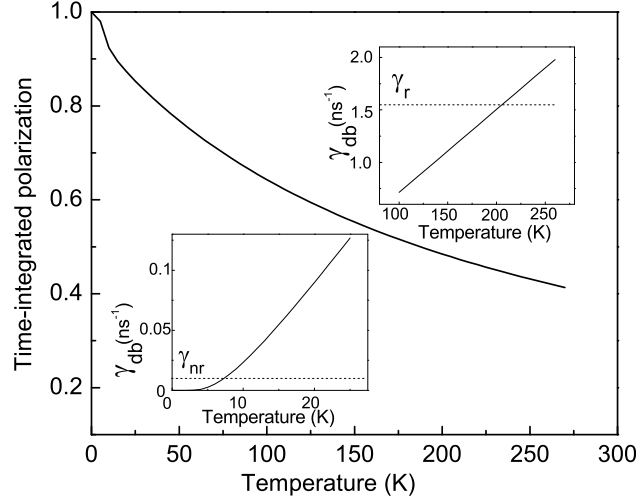


FIG. 2: Temperature dependence of the time-integrated polarization and the rate of a conversion of the dark to bright exciton at low (lower inset) and high (upper inset) temperatures in QDs from Ref.[13].

Using these data we extract the nonradiative relaxation rate to be $\gamma_{nr} \simeq 0.01 \text{ ns}^{-1}$. We are able also to calculate the rate γ_{db} at different temperatures and the result is presented in the insets in Fig.2. It can be seen that γ_{db} is one order of magnitude larger than γ_{nr} at $T \gtrsim 20$ K and becomes comparable (or even larger than) to γ_r at $T \gtrsim 200$ K. We can conclude therefore that at $T \gtrsim 20$ K the CdSe QDs examined in Ref.[13] are in a strong conversion regime and must show a noticeable decay of the linear polarization at high temperatures. To illustrate the expected depolarization effect, in Fig.2 we plot the integrated polarization $\overline{P} = \tilde{\gamma}_r(\tilde{\gamma}_r + \gamma_s)^{-1}$ (with γ_s calculated from (6)) in a wide temperature range up to room temperature. It can be seen that the values of \overline{P} are estimated to be close to unity at very low temperatures, but $\overline{P} \simeq 0.87$ is calculated already at 20 K and $\overline{P} \simeq 0.41$ is found at 270 K. The experimental proof of these theoretical results is certainly of interest.

c. Self-assembled In(Ga)As QDs in vertical field effect structures. In recent work [10] nonresonantly excited photoluminescence was measured from self-assembled InGaAs QDs in a vertical field effect structure. It was discovered that the rate γ_{db} of a conversion of the dark to bright exciton depends on the applied voltage and can be tuned to be smaller or larger than the recombination rate. It was found also that this rate increases with an increase of the temperature and the emission energy as well. These experimental findings

were successfully explained in Ref.[10] in the framework of a Kondo-like tunneling interaction between a localized electron in a dot and a delocalized electron in the back contact.

To draw a conclusion concerning the polarization dynamics in dots from Ref.[10], note that a strong dependence of the rate γ_{db} on a voltage rules out an intrinsic spin flip mechanism, as reported in Ref.[10]. It is evident also that an efficient tunneling mechanism including a simultaneous flip of the electron spin and the heavy-hole spin is unlikely, so that it is possible to suppose that still a conversion of the bright to dark exciton is faster than direct transitions between the bright states, as in the case of the spin relaxation of an intrinsic nature. Accordingly, the linear polarization dynamics in dots examined in Ref.[10] can be evaluated from the above expressions (4) and (5), where we again can chose $\gamma_L = 0$ [22]. Note also that for a tunneling mechanism the relation $\gamma_{bd} = \gamma_{db}e^{\Delta_0/kT}$ is still valid [10].

Table I. In(Ga)As QDs parameters after Ref.[10].

	dot1 (5 K)	dot2 (5 K)	dot2 (12 K)	dot2 (25 K)
E_{PL} (eV)	1.318	1.305	1.305	1.305
Δ_0 (meV)	0.3	0.6	0.6	0.6
τ_r (ns)	0.55	0.77	0.77	0.77
τ_{nr} (ns)	16.66	16.66	16.66	16.66
$\tau_{db, center}$ (ns)	120	110	20	2

The quantitative results obtained in Ref.[10] for two examined InGaAs dots differing by the exchange splitting Δ_0 , the radiative lifetime $\tau_r = \gamma_r^{-1}$ and the spin-flip time $\tau_{db} = \gamma_{db}^{-1}$, which we denote below as dot1 and dot2, are collected in Table I. The characteristic time $\tau_{nr} = \gamma_{nr}^{-1}$ and the values $\tau_{db, center}$ refer to the bias voltage applied in the center of the exciton plateau. The temperature at which the experiment was performed is shown in parentheses. In Fig.3 we plot the evolution of the linear polarization in dot1 for various voltages within the exciton plateau as shown in the inset. The curves are evaluated by means of the expression (4) with $\gamma_L = 0$. The involved rate γ_{db} is assumed to be $\gamma_{db} = \tau_{db, center}^{-1}$ for the curve V_c , $\gamma_{db} = \tau_{nr}^{-1}$ for the curve $V_{1nr, 2nr}$ and finally $\gamma_{db} = \tau_r^{-1}$ for the curve $V_{1r, 2r}$, so that all the necessary parameters are presented in Table I. In Fig.3 is seen that the polarization shows no decay on a scale of the radiative lifetime τ_r in the central part of the exciton plateau. However, the effect becomes considerable towards either the right or the left edge of the plateau (the curve $V_{1r, 2r}$), where the polarization decay time $\tau_s = \gamma_s^{-1} = 0.3$ ns (as estimated from (5) with $\gamma_L = 0$) is nearly two times smaller than $\tau_r = 0.55$ ns [23]. The respective

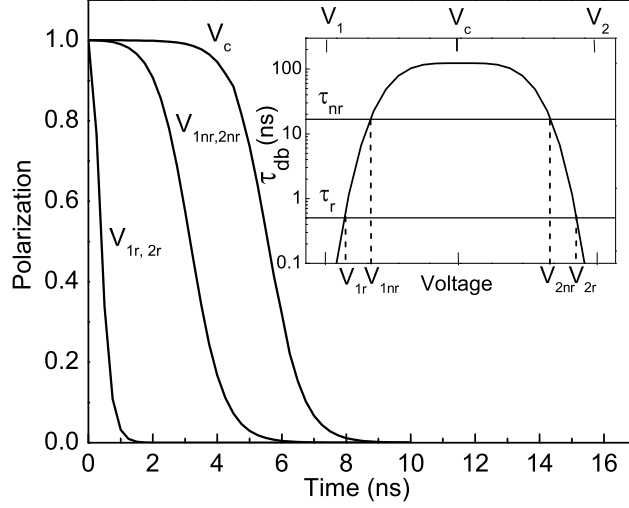


FIG. 3: The degree of polarization of the emission in the quantum dot from Ref.[10] as a function of time for various voltages as shown. The inset shows schematic dependence of the dark-to-bright relaxation time on a voltage within the neutral exciton plateau with the low-bias and the high-bias edge V_1 and V_2 , respectively, after Ref.[10].

value of the time-integrated polarization $\overline{P}_{\text{dot1}}(V_{1r,2r}) = 0.36$ markedly differs from unity. According to the experimental data presented in Table I for dot**2**, elevating temperature strongly accelerates the dark-bright transitions: the reported values of $\tau_{db, \text{center}}$ are two order of magnitude larger than τ_r at 5 K, but become comparable to τ_r at 25 K. Consequently, at moderate temperatures a noticeable effect on the polarization dynamics can be expected already in the center of the plateau. Calculating the time-integrated polarization from expression (5) with $\gamma_L = 0$ and using the data from Table I as input, we get $\overline{P}_{\text{dot2}}(V_c) = 0.997$ at 5 K, whereas $\overline{P}_{\text{dot2}}(V_c) = 0.71$ at 25 K.

In the context of excitonic spin relaxation in vertical field effect structures we would like to address Ref. [14]. Here the time-integrated polarization was investigated in individual InAs/GaAs self-assembled QDs. Experiments were performed at $T = 2$ K and the applied voltage was fixed at the value lying between the center and the high-bias edge of the neutral exciton plateau. The reported values of the characteristic time determining the time-integrated polarization for three examined isolated QDs (with a slightly different emission energy) range from 10 ns to 20 ns. These values are one order of magnitude longer than the radiative lifetime reported to be $\tau_r = 0.85$ ns [14]. Still they are too small to be

of an intrinsic nature, as we saw already at the beginning of this section. As an alternative version, the above discussed scenario of the voltage-controlled polarization dynamics can be proposed. This suggestion, of course, requires experimental verification.

In summary, the characteristic times of depolarization of the luminescence in strongly confined QDs are mainly controlled by a decay of the dark exciton and become comparable to the bright-to-dark relaxation times at negligible nonradiative losses. Numerical estimations with the relevant experimental data on the luminescence dynamics following nonresonant excitation demonstrate no depolarization effects in self-assembled InAs quantum dots at low temperatures. Similarly, the quantitative calculations indicate an efficient polarization decay in the high-quality self-assembled CdSe QDs at high temperatures and a voltage-dependent polarization dynamics in self-assembled In(Ga)As QDs in vertical field effect structures as well.

V. ACKNOWLEDGEMENTS

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